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**Abstract**

This project originated in an attempt to characterize the history of transport of radioactive contaminants in the Ob River, the fate of these contaminants within the river system, and their transport down the river to the estuary and Arctic Ocean. Our focus has been on the particle reactive contaminants $^{137}$Cs, $^{237}$Np, $^{238}$Pu, $^{239}$Pu, $^{240}$Pu and $^{210}$Pb. Expeditions were carried out to collect sediment cores from small lakes common on the Ob flood plain

The results may be summarized briefly as follows: 1) the sediments of the "sor" lakes of the flood plain do preserve records of the depositional history of the radioactive contaminants studied that extend to the pre-nuclear age. 2) All of the delta cores are similar in that they exhibit a strong maximum in both Pu isotopes and $^{137}$Cs at depth in the cores. 3) Based on $^{239,240}$Pu/Cs and $^{238}$Pu/$^{239,240}$Pu ratios, the chronology of the cores, and similarities between the Ob cores and one from the Taz River, a river with no nuclear facilities, we concluded that the Pu and $^{137}$Cs were derived from global fallout. 4) Uncertainties in our calculations would permit up to ~20% of these contaminants to be derived from non-fallout sources.

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## Arctic Radionuclides

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### Abstract

This project originated in an attempt to characterize the history of transport of radioactive contaminants in the Ob River, the fate of these contaminants within the river system, and their transport down the river to the estuary and Arctic Ocean. Our focus has been on the particle reactive contaminants $^{137}$Cs, $^{237}$Np, $^{238}$Pu, $^{239}$Pu, $^{240}$Pu and $^{210}$Pb. Expeditions were carried out to collect sediment cores from small lakes common on the Ob flood plain

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### INTRODUCTION

The drainage basin of the Ob River contains two of the major nuclear production plants of the former Soviet Union, Tomsk-7 and Mayak, at which large amounts of nuclear waste (~$1.5\times10^9$ Ci) are stored. Over their period of operation substantial amounts of radioactivity have been introduced to the environment as a result of both accidents and intentional releases. The history of releases in the past as well as the potential for catastrophic releases in the future has led to international interest in the fate and transport of nuclear contaminants in the Ob River and their delivery to the Arctic Ocean.

This project originated in an attempt to characterize the history of transport of radioactive contaminants in the Ob River, the fate of these contaminants within the river system, and their transport down the river to the estuary and Arctic Ocean. Our focus has been on the particle reactive contaminants $^{137}$Cs, $^{237}$Np, $^{238}$Pu, $^{239}$Pu, $^{240}$Pu as well as the naturally occurring isotope $^{210}$Pb as it is these isotopes that can leave a record of past transport in the sediments deposited from the river. Expeditions were carried out in the summers of 1994 (in the delta area) and 1995 (in the mid-reach of the river) to collect sediment cores from small lakes common on the Ob flood plain (Figure 1).

The initial results from analyses of these samples, presented previously at meetings and in papers, may be summarized briefly as follows: 1) the sediments of the "sor" lakes of the flood plain do preserve records of the depositional history of the radioactive contaminants studied, often with little or no mixing, that extend to the pre-nuclear age. 2) All of the delta cores are similar in that they exhibit a strong maximum

in both Pu isotopes and $^{137}$Cs at depth in the cores. 3) Based on $^{239,240}$Pu/Cs and $^{238}$Pu/$^{239,240}$Pu ratios, the chronology of the cores, and similarities between the Ob cores and one from the Taz River, a river with no nuclear facilities, we concluded that the Pu and $^{137}$Cs were derived from global fallout. 4) Uncertainties in our calculations would permit up to ~20% of these contaminants to be derived from non-fallout sources, but the data did not require additional sources.

## EVIDENCE FOR NON-FALLOUT SOURCES OF RADIOACTIVE CONTAMINANTS

Given that numerous releases of nuclear wastes from the plants had been documented, it was puzzling that we found no conclusive evidence of them in the delta sediments. This led us to look in more detail at isotopes that could provide a means of distinguishing between fallout and production plant sources of the nuclides of interest. In particular, through the auspices of Tom Beasley (US Dept. of Energy) and Jim Kelley (Battelle Northwest, Richland, WA, USA) mass spectrometric analyses of $^{239}$Pu, $^{240}$Pu, and $^{237}$Np were carried out on a sediment core from the Ob delta. In addition, we began analyses of the 1995 cores from the mid-reach of the Ob river and its major tributaries, the Irtysh and Tobol (cf. Figure 1). The data from these analyses provides clear evidence of non-fallout sources of radionuclides and indicate that there are at least three distinct sources of non-fallout nuclear contaminants to the rivers.

### $^{239}$Pu and $^{240}$Pu

The 239 and 240 mass isotopes of Pu provide a sensitive means of distinguishing between Pu derived from global fallout and material released from weapons production plants. Global fallout is characterized by a rather constant $^{240}$Pu/$^{239}$Pu ratio of 0.18, with the exception of the early 1950s. In contrast, ratios in weapons grade materials are around 0.04, values that are also observed in contaminated soils in the vicinity of the Mayak plant. The profile of $^{240}$Pu/$^{239}$Pu in a core from the Ob delta is presented in Figure 2. The $^{137}$Cs profile in the core is shown as a reference for fallout input. The latter shows the typical maximum in $^{137}$Cs which corresponds to the maximum in global fallout. The shoulder below the $^{137}$Cs maximum is, we believe, an expression of the bimodal fallout delivery created by the test series in the late fifties and then in the larger series in the early sixties. Also shown for reference is the expected fallout $^{240}$Pu/$^{239}$Pu ratio. Two aspects of the observed ratio are of particular importance. First, the ratio drops markedly in three distinct horizons. We interpret these variations as being due to the admixture of $^{239}$Pu-enriched material that cannot be of fallout origin. Rather, we ascribe each of these excursions to the admixture of weapons grade material released to the river. The decreases in $^{240}$Pu/$^{239}$Pu indicate addition of weapons grade material in amounts of 20 to 50% of the total Pu present. The restriction of these peaks to a single sample (1 cm) suggests to us that these are discrete events. Based on excess $^{210}$Pb chronology for the core these events occurred in approximately 1965, 1974, and 1991. The second aspect of these data that is important is the persistence of values below fallout. This suggests a chronic contamination with small amounts (~10%) of $^{239}$Pu-enriched material. The $^{239,240}$Pu/$^{137}$Cs ratios of the $^{239}$Pu-enriched samples are similar to fallout, whereas this ratio is much lower in sediments of the Techa River contaminated by the Mayak plant. The absence of decreases in the $^{239,240}$Pu/$^{137}$Cs ratio in core 7B indicates that Mayak is not the source. Further, The $^{240}$Pu/$^{239}$Pu ratio of particulate matter collected in the rivers on the 1995 expedition (data from Cochran and Moran, pers comm.) demonstrates that only the Ob above the confluence with the Irtysh has the low ratios characteristic of the delta sediment profile of Figure 2, pointing to a likely upstream source from Tomsk-7.

### $^{237}$Np/$^{239}$Pu ratios

Neptunium ($^{237}$Np) is also present in global fallout, typically having the ratio to $^{239}$Pu ($^{237}$Np/$^{239}$Pu) of ~0.45. Analyses of the Ob94-7B sediment core present clear evidence of a second and very different

source of non-fallout material in the Ob River basin (Figure 3). The $^{237}Np/^{239}Pu$ ratio below about 6 cm is essentially that expected for fallout. The one exception (14-15 cm) occurs in the deepest of the $^{239}Pu$-enriched horizons and is the result of presenting the data in this fashion. Above 6 cm the ratio increases rapidly to almost 3 times the fallout value, indicating the introduction of $^{237}Np$ to the river system. It appears that elevated ratios persisted at least up until the collection of the core in 1994. The 2-3 cm horizon is another of the $^{239}Pu$-enriched layers and hence the sharp drop here is, as noted above, an artifact of presenting the data as a ratio. Based on excess $^{210}Pb$ chronology, non-fallout values were first deposited around 1983, peaked around 1987, and fell somewhat to the core top value, one still almost twice fallout. The $^{237}Np$ increase is not synchronous with the deposition of fallout $^{137}Cs$, nor does it appear to be related to the $^{239}Pu$-enriched layers, rather, it appears to be from a completely different source. The $^{237}Np/^{239}Pu$ ratios of particulate matter vary substantially with many samples being below fallout values. Only samples from the Irtysh above the Tobol have high ratios, values that are very similar to those in the core top (e.g. ~0.8), suggesting a source on this river. While geochemical fractionation may influence the chronology of the $^{237}Np/^{239}Pu$ ratios, we believe a source within the basin is likely, but its identity remains unknown to us.

### $^{137}$Cs: "non-global" fallout

The third non-fallout source of radionuclide contamination identified in the Ob basin appears to have introduced $^{137}Cs$. The cores in the delta exhibit small increases in $^{137}Cs$ at a few depths above the main fallout maximum (cf. Figure 2 at 3-4 and 8-9 cm) . Analyses of $^{137}Cs$ in the 1995 cores from the mid-reach of the river system present a very different picture. The sediments of this area are often deposited at substantially higher rates and hence the length scale is more drawn out and the depth to what we identify with the global fallout deposition maximum is greater, by as much as a factor of 4. In these sediments, in addition to the relatively deep global fallout feature, there is a strong, shallow secondary $^{137}Cs$ maximum (Figure 4). Unlike the global fallout maximum in which there is a corresponding Pu maximum, the shallow increase in $^{137}Cs$ is not accompanied by an increase in $^{239,240}Pu$ (10-13 cm, Figure 4). Thus this cannot be attributed to an influx of fallout $^{137}Cs$, but rather a source much lower in Pu than fallout is required. A second feature of importance in identifying the source of the $^{137}Cs$ lies in the fact that the shallow strong maximum is found in sediments from both the Tobol (likely to receive Mayak contaminants) and Irtysh (no nuclear plants) rivers. Thus the source must be capable of depositing material in these separated and distinct drainage basins. Finally, the chronology of the cores places deposition of the shallow $^{137}Cs$ peak at around 1985. On the basis of these characteristics, we have concluded that the source of the shallow $^{137}Cs$ peak is, primarily, fallout from the Chernobyl accident.

## CONCLUSIONS

The analysis of plutonium and neptunium isotopes and $^{137}Cs$ in sediments from the lakes of the Ob, Irtysh, and Tobol flood plains lead us to the following conclusions regarding sources of these nuclear contaminants to the river:

1) $^{239,240}Pu$ and $^{137}Cs$ in the sediments of the Ob River are primarily derived from global fallout. There is, however, clear evidence of three non-fallout sources of nuclear contaminants to the river, in addition to the $^{129}I$.
2) Based on distributions of $^{239}Pu$ and $^{240}Pu$, material with the characteristics of weapons grade plutonium has been transported in the river and deposited in narrow horizons on at least three occasions. Within these layers the non-fallout component can be roughly as abundant as fallout Pu. The most likely source of this material is the upper Ob River and the Tomsk-7 plant located near it.
3) $^{237}Np$ distributions in the sediments of the delta indicate that since about 1983 this isotope has been transported and deposited in ratios quite distinct from fallout and unrelated to the events characterized

by $^{239}Pu$ enrichments (2 above). We have not, as yet, identified the source nor ruled out the influence of geochemical fractionation, but evidence indicates that the high ratios emanate from the Irtysh River.

4) In addition to fallout $^{137}Cs$, deposition of this isotope in the mid 1980s indicates that the fallout from Chernobyl has been deposited in the southerly rivers of the Ob basin.

## ACKNOWLEDGMENTS

This research on the nuclear contamination of the Ob River could not have taken place without the dedication, skill, and determination of our colleague Gera Pantaleyev. Gera led the 1994 expedition and was in the process of leading the 1995 expedition when he lost his life in an accident on the Ob River. He is sorely missed by all who knew him. Financial support for this research has been provided through the US Navy, Office of Naval Research, Arctic Nuclear Waste Assessment Program.

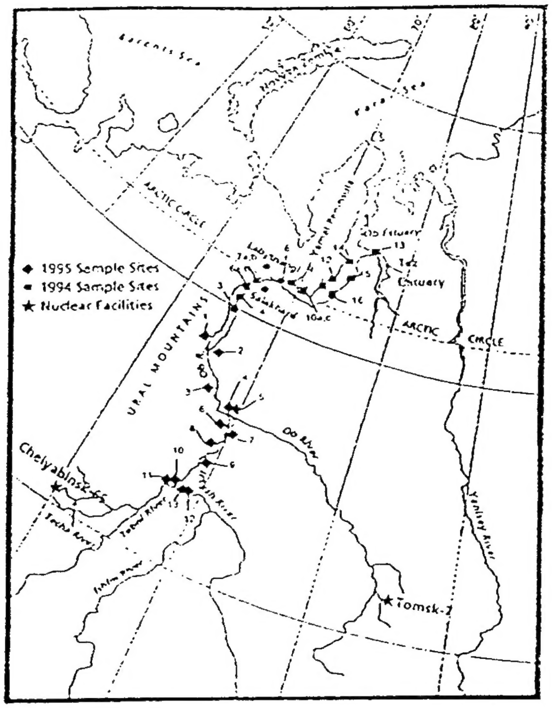


Figure 1- Locations of sediment cores collected on the Ob, Irtysh, and Tobol Rivers during expeditions in 1994 and 1995.

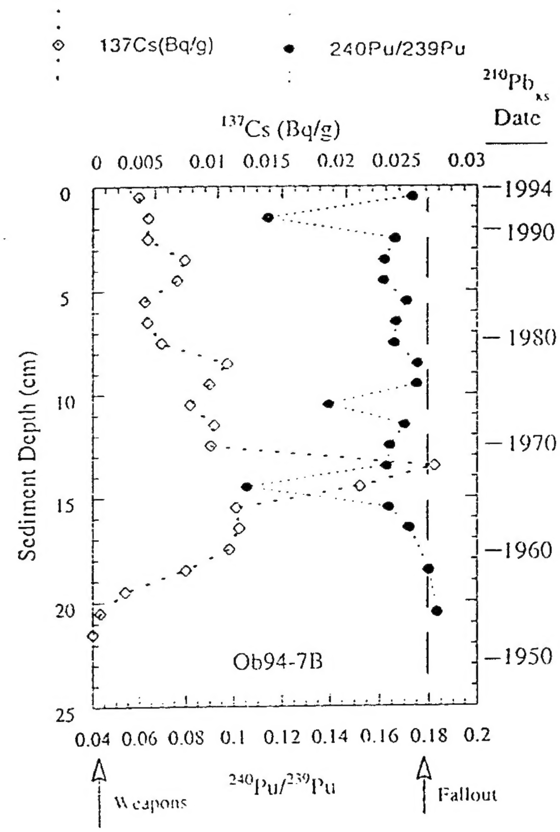


Figure 2- Distributions of $^{137}$Cs and $^{240}$Pu/$^{239}$Pu ratio in a sediment core from the Ob delta. The $^{137}$Cs is shown to illustrate the record and timing of deposition from global fallout. Also shown for reference are the $^{240}$Pu/$^{239}$Pu ratios of global fallout an d typical weapons grade material. The time scale on the right is based on the average accumulation rate determined from excess $^{210}$Pb.

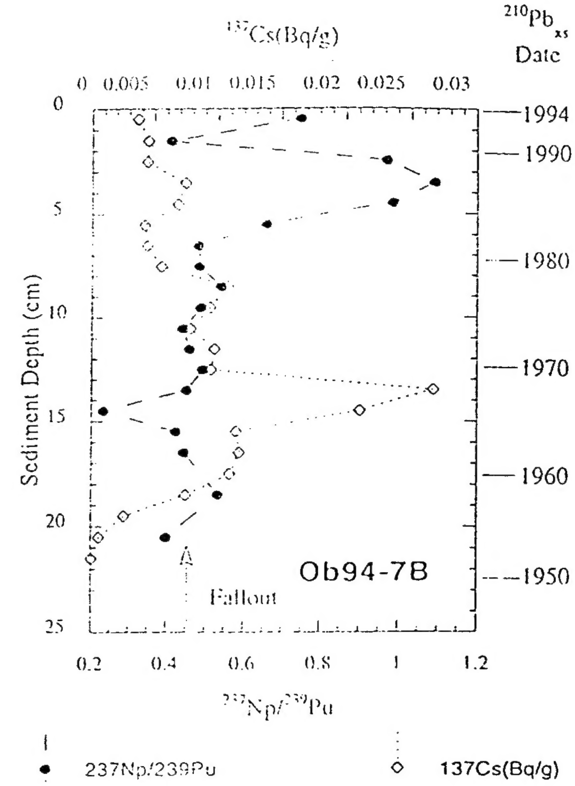


Figure 3- The distribution of $^{137}$Cs and the $^{237}$Np/$^{239}$Pu ratio in a sediment core from the Ob delta (same as Figure 2). See Figure 2 caption.

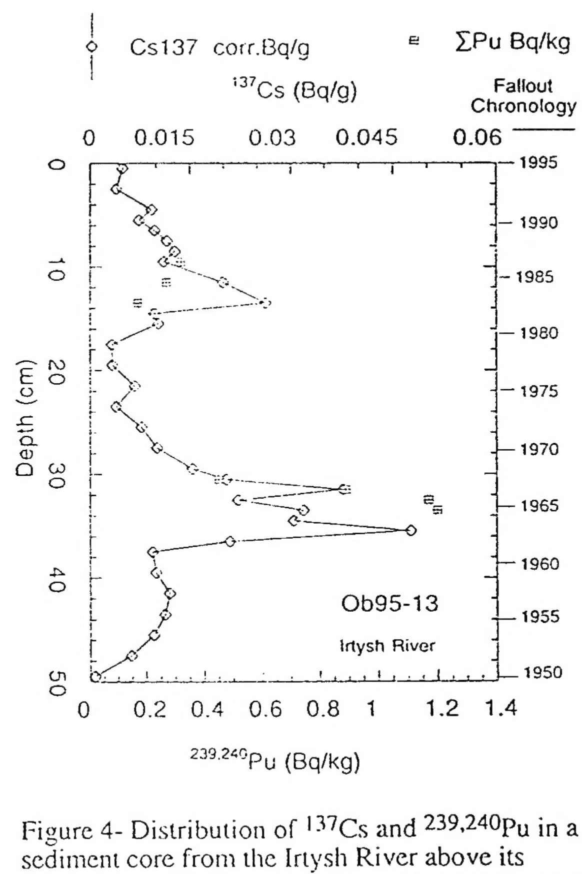


Figure 4- Distribution of $^{137}$Cs and $^{239,240}$Pu in a sediment core from the Irtysh River above its confluence with the Tobol. Chronology is based on an average accumulation rate estimated from assigning a date of 1965 to the fallout maximum and 1950 to the introduction of $^{137}$Cs.